



## Project Summary

# Field Measurement of Greenhouse Gas Emission Rates and Development of Emission Factors for Wastewater Treatment

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A research project was conducted to measure emissions of greenhouse gases (GHGs) from wastewater treatment (WWT) and disposal facilities. The overall objective of the research under the base statement of work area for this contract is to develop more reliable estimates of GHG emissions from industrial and domestic WWT systems. Most previous research for these sources has used a mass balance approach to estimate potential methane ( $\text{CH}_4$ ) emissions, but in this study emissions of  $\text{CH}_4$  and other GHGs were measured under field conditions, which should improve the reliability of the emission estimates.

Field sampling was performed at five sites, including WWT systems in the beef and chicken processing industries and two publicly owned treatment works (POTWs). Ambient air was measured immediately downwind of the lagoons using a Fourier Transform Infrared (FTIR) approach. The FTIR light beam was directed along a path of several hundred feet and the absorbance of gases was measured. The target compounds of interest included  $\text{CH}_4$ , carbon dioxide ( $\text{CO}_2$ ), nitrous oxide ( $\text{N}_2\text{O}$ ), carbon monoxide ( $\text{CO}$ ), ammonia ( $\text{NH}_3$ ), and certain volatile organic compounds (VOCs). The source term (i.e., emission rate) can be determined from information about the average downwind ambient concentration (measured by the FTIR method) and the atmospheric dispersion characteristics at the time of sampling. In addition, samples of influent and effluent wastewater and sludge were collected. Emission

factors were developed in terms of grams of species emitted per gram of precursor in the influent wastewater; e.g.,  $\text{g CH}_4/\text{g}$  biological oxygen demand (BOD). The emission factors will be combined with activity factor data to develop national and global emissions inventories.

*This Project Summary was developed by EPA's National Risk Management Research Laboratory's Air Pollution Prevention and Control Division, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back.)*

### Introduction

A GHG generally can be defined as any molecule which absorbs infrared light in the spectral region of 5 to 20  $\mu\text{m}$ . These molecules include water vapor ( $\text{H}_2\text{O}$ ),  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ , certain VOCs, and  $\text{N}_2\text{O}$ . Reasonably accurate global balances of GHGs are needed as input to climatic models for estimating long-term global temperature changes.

A large number of natural and anthropic activities produce or release GHGs. The emphasis of this program was on emissions from WWT facilities. The decomposition of organic waste may occur aerobically (i.e., with oxygen) or anaerobically (i.e., without oxygen). Aerobic decomposition of organic carbon results in the production of  $\text{CO}_2$ , while anaerobic decomposition results in the production of  $\text{CH}_4$  and  $\text{CO}_2$ . Given a sufficient amount of time, essentially every atom of carbon in waste streams is converted to either  $\text{CO}_2$  or  $\text{CH}_4$ .

In terms of their ability to retain heat in the atmosphere, however,  $\text{CO}_2$  and  $\text{CH}_4$  are not equivalent. A given mass of  $\text{CH}_4$  is 58 times stronger a GHG than the same mass of  $\text{CO}_2$  (it is 21 times stronger on a molecular basis). Therefore, the relative amount of anaerobic versus aerobic decomposition is of interest.

The overall objective of the research under the base statement of work area for this contract was to develop more reliable estimates of GHG emissions from industrial and domestic WWT systems. National and global inventories of  $\text{CH}_4$  emissions from WWT facilities have been published. These estimates, however, are based on various assumptions and very limited data. Most previous research for these sources has used a mass balance approach to estimate potential  $\text{CH}_4$  emissions, but in this study emissions of  $\text{CH}_4$  and other GHGs were measured under field conditions, which should improve the reliability of the emission estimates.

The overall objectives of this study were: 1) Identify industries and WWT processes that have the greatest potential for measurable emissions of  $\text{CH}_4$ ; 2) Select the most promising sites for testing in those industries; 3) Perform ambient air measurements using an open path monitoring (OPM) approach with a FTIR spectroscopy instrument; 4) Collect and process data and characterize the influent and effluent wastewater quality at the field sites; and 5) Use the field data to develop emission factors for each target compound. The target compounds of interest included  $\text{CH}_4$ ,  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{CO}$ ,  $\text{NH}_3$ , and certain VOCs.

A subset of WWT systems that employ anaerobic treatment processes was selected for testing. Within this subset, anaerobic lagoons were given priority over anaerobic digesters, tanks, and sludge disposal units because lagoons offered the fewest logistical constraints to testing. Although anaerobic lagoons are not extensively used to treat industrial and domestic waste in the U.S., other countries use anaerobic lagoons to treat wastewater. Because of difficulties associated with identifying sites and the expense of conducting field measurements in foreign countries, sites in the U.S. that are representative of treatment conditions in developing countries were selected for testing.

## Approach

Site selection focused on U.S. WWT systems that employ open, anaerobic processes to achieve high levels of BOD or chemical oxygen demand (COD) removal. First, industries that treat large volumes of wastewater and remove large amounts of

BOD/COD were identified. Approximately a dozen industries were identified as potential candidates for testing. Second, a telephone survey was conducted of industry representatives, experts in the WWT area, and regulatory personnel to identify industries most likely to treat wastewater to remove high levels of BOD/COD in open, anaerobic lagoons. The most promising candidates were beef and poultry processing plants, and pulp and paper mills. POTWs also were of interest because they are used to treat a significant fraction of wastewater both nationally and globally and, also, are thought to be a potentially significant source of  $\text{N}_2\text{O}$  emissions.

Five sites were selected for testing. The selection intentionally included sites from several different industries: two beef processing plants, one chicken processing plant, and two POTWs. Two sites from certain industries were included to help determine the variability in emissions within a given industry.

The field work involved being on site for about 5 days at each facility. Ambient air was measured immediately upwind and downwind of the lagoons using an OPM-transect method (OPM-TM) approach with detection by FTIR spectroscopy. The FTIR light beam was directed along a path of several hundred feet and the absorbance of gases was measured. Emission rates were determined from measurements of the downwind ambient concentration and the atmospheric dispersion characteristics at the time of sampling. In addition, a limited number of influent and effluent wastewater and sludge samples were collected.

Emission factors were developed in terms of grams of GHG species emitted per gram of precursor in the influent wastewater (e.g., g  $\text{CH}_4$ /g BOD). The emission factors will be combined with activity factor data to develop national and global emission estimates.

## Results

OPM-TM using the FTIR was used to determine emission rates. A very large data set was generated, and up to 300 separate valid, 5-minute average emission rate determinations were made at a given site. The air measurement data were reviewed to identify compounds found in significantly greater concentrations in the downwind air versus the upwind air at each site. Any such compounds were likely to have been emitted from the lagoons being tested. Many of the target analytes were found at the same concentration levels upwind and downwind of the lagoons; i.e., they had no quantifiable emission rate.

Only  $\text{CH}_4$ ,  $\text{NH}_3$ , and the sulfur hexafluoride ( $\text{SF}_6$ ) tracer gas generally were present in greater amounts in the downwind air.

The minimum quantifiable emission rate varied from site to site and from one 5-minute period to another. The detection limit for a given compound, in terms of grams per second, is dependent on the smallest difference between downwind and upwind concentrations that could be identified apart from the measurement variability within each of the upwind and downwind data sets. For each increment of 0.5 ppmv (500 ppbv) that a given compound was present in greater concentrations downwind than upwind, its emission rate was about 1 g/sec (depending on the molecular weight of the compound). Typical detection limits were about 0.1 g/sec for most compounds, except for  $\text{CO}_2$ , which had a minimum detection limit of about 150 g/sec. The high detection limit for  $\text{CO}_2$  was due to the high background concentrations (e.g., 500 ppmv) and the measurement coefficient of variability (e.g., CV = 7.5%, or 37.5 ppmv).

At all three meat processing plants, large amounts of  $\text{CH}_4$  were detected downwind of the WWT system. For the two beef processing plants, the concentration of  $\text{CH}_4$  (and  $\text{NH}_3$ ) exhibited an exponential relationship with wind speed. The downwind  $\text{CH}_4$  concentration at the chicken processing plant did not show a clear relationship between concentration and wind speed. At the chicken processing plant, however, the range of wind speeds was much smaller than for the meat processing plants and the number of valid measurement periods also was much smaller, making it more difficult to identify trends and relationships. There also was a thick grease layer present on top of the lagoon which would tend to diminish the effect of surface winds on air emissions.

The emission rates measured at each site for  $\text{CH}_4$ ,  $\text{NH}_3$ , and other selected compounds are given in Table 1. Surprisingly, no emissions were detected from the POTWs. It was expected that either  $\text{CH}_4$  or  $\text{CO}_2$  would be detected. The dissolved oxygen (DO) level in the lagoons exceeds 2 mg/L, indicating that BOD removal is taking place under aerobic conditions. So it is highly probable that  $\text{CO}_2$  is being generated, but the levels were too small to detect given the very high background levels of  $\text{CO}_2$  and the measurement variability. In general, anaerobic degradation can be expected to produce a mixture of  $\text{CH}_4$  and  $\text{CO}_2$  (somewhere between a 50:50 and a 70:30 ratio). Therefore, emissions of  $\text{CO}_2$  would be expected wherever quantifiable emission rates of  $\text{CH}_4$  were found.

**Table 1.** Measured Emission Rates of Selected Compounds for Each Field Site

Site	Compound	Average Downwind Conc. (ppm)	Average Upwind Conc. (ppm)	Maximum Downwind Conc. (ppm)	Average Emission Rate (g/sec)
Beef Processing Plant in SW U.S.	CH <sub>4</sub>	61.9	2.3	142	280
	NH <sub>3</sub>	355 ppb	0	609 ppb	2.2
Beef Processing Plant in Midwest U.S.	CH <sub>4</sub>	58.1	2.83	200	230
	NH <sub>3</sub>	1.04	0.277	2.06	3.5
Chicken Processing Plant in SE U.S.	CH <sub>4</sub>	9.80	1.92	29.9	180
	NH <sub>3</sub>	2.6 ppb	2.8 ppb	44.1 ppb	0.066
	N <sub>2</sub> O	563 ppb	542 ppb	586 ppb	2.6
POTW for Small Town in SW U.S. <sup>a</sup>	CH <sub>4</sub>	2.20	2.14	2.46	<0.15
	NH <sub>3</sub>	0.2 ppb	0	15.4 ppb	<0.05
	CO <sub>2</sub>	342	351	384	<150
POTW for Very Small Town in SW U.S. <sup>a</sup>	CH <sub>4</sub>	2.11	2.16	2.81	<0.15
	NH <sub>3</sub>	93.3 ppb	25.5 ppb	214 ppb	<0.05
	CO <sub>2</sub>	528	668	691	<150

<sup>a</sup>Methane, carbon dioxide, and ammonia values are shown for the POTWs for comparison purposes. No quantifiable emissions of these compounds were detected at either POTW.

The lack of quantifiable CO<sub>2</sub> emission rates may be due to the high detection limit for CO<sub>2</sub> emission rates, as previously discussed. The absence of CO<sub>2</sub> emissions also could be due to the presence of cyanobacteria (blue-green algae) in the anaerobic lagoons.

The wastewater data for all three meat processing plants are very similar, with the two beef processing plants showing very good agreement. All three WWT systems have high BOD removal rates (88-95%), as well as high removal rates for COD, total organic carbon (TOC), and nitrates. All three WWT systems at meat processing plants generated large amounts of NH<sub>3</sub> as a by-product of the biodegradation of the wastewater. The only parameter that showed variable behavior from system to system was total Kjeldahl nitrogen (TKN).

The two POTWs had similar influent wastewater and exhibited similar performance in terms of removal of BOD, COD, TOC, TKN, and NH<sub>3</sub>. Both systems generated nitrates as a by-product of biodegradation.

Activity factors were developed for each site based on information provided by the plant operators and from the wastewater data. Emission factors were developed for each site by dividing the average emission rates by the activity factors for each site. The resulting emission factors are given in Table 2. For CH<sub>4</sub>, the emission factor based on COD should be a better

predictor of emissions from other facilities than the emission factor based on BOD. The 5-day BOD test will not fully degrade all of the biological material in wastewaters containing proteins and fatty acids. The suspended solids associated with the wastewaters also are biodegradable, and

their ultimate BOD would not be exerted in the 5 days it takes to run a standard BOD test. COD data, however, are not always available, and estimates based on other activity factors may be necessary. Therefore, a variety of emission factors are included in Table 2.

**Table 2.** Average Emission Factors

Compound	Emission Factor	Average	Range
Methane	g CH <sub>4</sub> /head of cattle	4,200	3,500 - 4,800
	g CH <sub>4</sub> /chicken	120	N/A
	g CH <sub>4</sub> /kg meat	37	15 - 74
	g CH <sub>4</sub> /L of wastewater	2.7	1.6 - 4.6
	g CH <sub>4</sub> /g influent BOD	1.5	0.40 - 3.2
	g CH <sub>4</sub> /g BOD removed	1.6	0.43 - 3.4
	g CH <sub>4</sub> /g COD removed	0.96	0.26 - 2.0
Ammonia	g NH <sub>3</sub> /head of cattle	46	37 - 54
	g NH <sub>3</sub> /chicken	0.046	N/A
	g NH <sub>3</sub> /kg meat	0.14	0.027 - 0.24
	g NH <sub>3</sub> /L of wastewater	0.014	0.0017 - 0.028
	g NH <sub>3</sub> /g influent BOD	0.40	0.0031 - 1.2
	g NH <sub>3</sub> /g NH <sub>3</sub> in effluent	0.072	0.020 - 0.13
Nitrous Oxide	g N <sub>2</sub> O/chicken	1.8	N/A
	g N <sub>2</sub> O/kg meat	1.1	N/A
	g N <sub>2</sub> O/L of wastewater	0.067	N/A
	g N <sub>2</sub> O/g BOD removed	0.051	N/A
	g TKN removed	1.7	N/A

N/A = Not applicable.

An estimate of the uncertainty of the emission factors was developed through standard error propagation methods. The derived emission factors all appear to be reliable to within a factor of 2, based on random error in the measurements, and assuming that the sites and samples accurately represent the population of interest.

It is possible that the lagoons are a sink for suspended and colloidal material (i.e., insoluble BOD) and this material builds up over time in the lagoon sediments. If so, sediments may degrade during summer months or whenever the sediment is re-suspended, thereby increasing the CH<sub>4</sub> (and CO<sub>2</sub>) emissions. However, no seasonal trend is evident in the BOD effluent levels in the long-term wastewater data provided by the plants.

A number of previously published studies contain estimated or measured values for the emission fluxes of CH<sub>4</sub> from liquid surfaces or slurries. The key comparison is the emission flux (i.e., emission rate per unit surface area). The average CH<sub>4</sub> emission flux for the three meat processing plants ranged from 6,100 to 23,000 µg

CH<sub>4</sub>/sec-m<sup>2</sup>. Results for livestock lagoons in previous studies (1,400 to 9,400), were within an order of magnitude, as were measurements at a manure tank (1,300 to 3,800). The emission flux from municipal WWT systems, industrial WWT systems, and rice paddies was substantially lower, as expected given the much lower BOD and COD levels in such waters.

Very few published emission factors can be compared with the emission factors developed in this study. The most widely reported emission factor for CH<sub>4</sub> is 0.22 g CH<sub>4</sub>/g BOD. The reference for this factor does not provide information about how it was developed. It is very close to the theoretical value for the anaerobic degradation of glucose. The emission factors determined in this study are substantially higher than those based on glucose degradation. Glucose is a simple sugar and its biodegradation over short periods of time cannot be directly compared with the microbial degradation of complex mixtures of amino and fatty acids, such as are present in the wastewaters at the meat processing plants.

## Conclusions

Several conclusions can be drawn from the study:

- The FTIR measurement approach used in this study was successful for the simultaneous collection of large amounts of ambient concentration data for CH<sub>4</sub> and NH<sub>3</sub>;
- The use of the OPM-TM approach using FTIR for estimating emission rates has insufficient sensitivity for certain compounds, such as hydrogen sulfide and total non-methane hydrocarbons, due to limitations in the FTIR analysis. For most of the sites, the sensitivity for CO<sub>2</sub> was limited by the high background concentrations and the variability in the background concentrations;
- Anaerobic WWT lagoons are a significant source of CH<sub>4</sub> and NH<sub>3</sub> emissions; and
- Lagoons at POTWs are not a significant source of any GHG, with the possible exception of CO<sub>2</sub>.

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*The complete report, entitled "Field Measurement of Greenhouse Gas Emission Rates and Development of Emission Factors for Wastewater Treatment," (Order No. PB98-117898; Cost: \$67.00, subject to change) will be available only from:*

*National Technical Information Service*

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